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Lung deposited surface area size distributions of particulate matter in different urban areas

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HIGHLIGHTS

- Lung deposited surface area (LDSA) size distributions measured in a metropolitan area.
- An electrical low pressure impactor (ELPI) calibrated to measure the LDSA.
- The LDSA was influeced by traffic more than the mass of fine particles (PM2.5).
- Both the nucleation and soot mode found to have a contribution to the LDSA.
- Size distribution data is important for the use of epidemiologial studies.

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ABSTRACT

Lung deposited surface area (LDSA) concentration is considered as a relevant metric for the negative health effects of aerosol particles. We report for the first time the size distributions of the LDSA measured in urban air. The measurements were carried out in the metropolitan area of Helsinki, including mobile laboratory and stationary measurements in different outdoor environments, such as traffic sites, a park area, the city center and residential areas. The main instrument in this study was an electrical low pressure impactor (ELPI), which was calibrated in the field to measure the LDSA concentration. The calibration factor was determined to be 60 $\mu m^2/(cm^3\ pA)$. In the experiments, the LDSA size distributions were found to form two modes at the traffic sites and in the city center. Both of these traffic related particle modes, the nucleation mode and the soot mode, had a clear contribution to the total LDSA concentration. The average total concentrations varied from 12 to 94 $\mu m^2/cm^3$, measured in the park area and at the traffic site next to a major road, respectively. The LDSA concentration was found to correlate with the mass of fine particles (PM2.5), but the relation of these two metrics varied between different environments, emphasizing the influence of traffic on the LDSA. The results of this study provide valuable information on the total concentrations and size distributions of the LDSA for epidemiological studies. The size distributions are especially important in estimating the contribution of outdoor concentrations on the concentrations inside buildings and vehicles through size-dependent penetration factors.

1. Introduction

Particulate matter in urban air is a significant risk to human health. Over the past few decades, this has been taken into account in the legislation controlling the urban air quality and emission sources. Still, the particulate matter is estimated to cause worldwide about 2.1 million deaths per year (Silva et al., 2013) and contribute to the incidence of various cardiopulmonary diseases and lung cancer (Pope III et al., 2002; Hoek et al., 2002). The first epidemiological evidence of the inverse health effects of urban aerosols was based on particle mass concentration measurements

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(Dockery et al., 1993). After that, the particulate matter in urban air has been widely monitored, reported and restricted by legislation. Both the mass of respirable suspended particles (PM10) and the mass of fine particles (PM2.5) are included in the standardized outdoor air quality measurement, but only the mass of fine particles has been associated to premature deaths and cardiopulmonary diseases (Silva et al., 2013).

Number concentration and number size distribution measurements of fine particles have had an important role in understanding the climatic effects of aerosols, such as cloud formation. In urban air, these have been reported in various studies (Shi et al., 1999; Virtanen et al., 2006; Pirjola et al., 2006, 2012; Pant and Harrison, 2013) and the aerosols originated from anthropogenic sources have been estimated to contribute significantly to global climate (Schwartz, 1996; Rotstayn et al., 2009). Urban aerosols are often traffic-related and their particle number size distributions consist of two modes, i.e. a nucleation mode and a soot mode, the latter of which can also be referred as an accumulation mode in urban air. Because of the legislative changes and the development of engine technologies, the relation of these two modes is changing. The role of the number concentration and number size distribution measurements is important in characterizing the particulate emissions of different engine and after treatment technologies. However, there are no clear epidemiological nor toxicological evidence on the correlation of the inverse health effects and the number concentration of fine particles (HEI Review Panel on Ultrafine Particles, 2013).

Despite of the strong epidemiological evidence for the inverse health effects of PM2.5, the mechanisms affecting human health are still poorly understood. Harmful particles are most probably entering human circulation through the alveolar region of lungs and the inverse health effects can be associated to the surface chemistry of the particles. It has been proposed that the chemical effect of particles could be strictly measured by observing the reactive oxygen species of aerosols (Stevanovic et al., 2013). However, this method is based on off-line analyses of collected samples and it is not applicable for on-line monitoring. A simple manner to combine the lung deposition of particles and the potential for the surface chemistry is to use a metric called the lung deposited surface area (LDSA) concentration. The surface area of particles has been shown to correlate with inverse health effects in toxicological studies (Brown et al., 2001; Oberdörster et al., 2005).

The LDSA is closely related to the concepts of the condensation sink and ion sink, describing the mass transfer rate of molecules or ions onto the particles. Thus, the LDSA concentration strongly correlates with the ion attachment and diffusion charging, and it is relatively easy to measure with diffusion charger based instruments. One of these instruments is a nanoparticle surface area monitor (NSAM) (Fissan et al., 2007) which is enable to measure the lung deposited surface area concentrations for both alveolar and tracheobronchial regions of lungs. Recently, several miniaturized instruments, such as a miniDiSC (Fierz et al., 2011), NanoTracer (Marra, 2011) and a Partector (Fierz et al., 2014), have been introduced to measure the LDSA concentration. All of these instruments provide information on the total LDSA concentration but not the size distribution. There are instruments such as an electrical low pressure impactor (ELPI, Dekati Ltd.) (Keskinen et al., 1992) and a cascade epiphaniometer (CEPI) (Gäggeler et al., 1989) that are in principle capable of measuring surface related quantities as a function of the particle size. Previously, Kuuluvainen et al. (2010) used an ELPI to measure condensation sink and ion sink, and Gini et al. (2013) used a CEPI to measure active surface size distributions.

The LDSA concentrations have been reported for indoor aerosols during different activities (Mokhtar et al., 2013) and aerosols emitted from a nanoparticle manufacturing process (Mäkelä et al., 2009). Ambient measurements have been conducted at urban background sites in Barcelona (Reche et al., 2015), Minneapolis (Wilson et al., 2007), Zürich (Fierz et al., 2011), Los Angeles (Ntziachristos et al., 2007) and Lisbon (Gomes et al., 2012) having the LDSA total concentrations between 10 and 89 $\mu m^2/cm^3$ Ntziachristos et al. (2007) reported the LDSA concentrations also at traffic sites in Los Angeles ranging from 106 to 153 $\mu m^2/cm^3$. The amount of the reported total LDSA concentrations for different environments found in the literature is not extensive. More data is needed especially for the use of exposure estimation and epidemiological studies (Buonanno et al., 2011). The size distributions of the LDSA, which have not been reported in any previous studies, could be useful to estimate the contribution of outdoor concentrations on the concentrations inside buildings and vehicles. Chen and Zhao (2011) reviewed the relationship between indoor and outdoor particle concentrations, and the size-dependency of the penetration and infiltration was emphasized.

In this study, we report the lung deposited surface area size distributions of particulate matter in different urban outdoor environments of the metropolitan area of Helsinki. The results include a large amount of data from traffic sites, a park area, the city center and residential areas. The role of the LDSA size distributions is discussed with respect to other metrics and requirements of epidemiological studies.

2. Experimental

2.1. Aerosol instruments

The main aerosol instrument used in this study was an electrical low pressure impactor (ELPI, Dekati Ltd.). The operation principle and the mechanical structure of the ELPI was introduced by Keskinen et al. (1992) and later described in detail by Marjamäki et al. (2000). The instrument consists of a unipolar corona charger and a cascade impactor where particles are classified into 13 stages according to their aerodynamic size. Twelve of these stages are connected to electrometers in order to measure the electric charge carried by the particles. In this study, the original configuration of the ELPI was modified by a filter stage (Marjamäki et al., 2002) and an additional impactor stage (Yli-Ojanperä et al., 2010). Both of these modifications enhance the size resolution of the ELPI at the particle size range below 30 nm. Apiezon-L vacuum grease was used on the collection substrates of the impactor in order to prevent the bounce of the collected particles. The bounce has been characterized recently in the ELPI (Kuuluvainen et al., 2013) and with respect to fundamental parameters (Arffman et al., 2015; Rennecke and Weber, 2013). In the typical use of the ELPI, the electric current is used to determine the particle number concentration and number size distribution. Because the charging of the particles in the ELPI is mainly based on diffusion charging, the response of the instrument can be used more directly to determine surface related quantities. In this study, after a simple calibration procedure described below, we calculated the LDSA size distributions at the particle size range from 7 nm to 10 μm.

Also the operation of a nanoparticle surface area monitor (NSAM, TSI Inc., Model 3550) is based on the electrical charging of particles and electric current measurement. The NSAM is equipped with a Faraday-cup filter which is used to measure the electric current from the collected particles. The total LDSA concentration is calculated from the electric current as described by Fissan et al. (2007) and Asbach et al. (2009). The NSAM has two operational modes, the alveolar and tracheobronchial deposition modes, with different ion trap voltages. In this study, the NSAM was used in the alveolar deposition mode with a standard 1 μm pre-cut cyclone. The NSAM was the primary reference instrument used in the

calibration of the ELPI. In addition, a differential mobility particle sizer (DMPS), consisting of a differential mobility analyzer (DMA, Vienna type) and a condensation particle counter (CPC, Grimm Model 5.401), was used to particle number size distribution measurement and a tapered element oscillating microbalance technique (TEOM, Thermo Scientic, Model 1400AB) was used for PM2.5 measurements.

2.2. Stationary measurements

Stationary measurement campaigns were performed at five different urban environments in the metropolitan area of Helsinki. Table 1 shows the location, time period and short description of each campaign. In addition, the range of basic weather parameters, such as the temperature and relative humidity, are shown. The duration of each measurement campaign was approximately two weeks. The measurements at the traffic site near the major road and in the city center were conducted simultaneously, as well as the measurements in the suburban residential area and in the residential area in the inner city. The measurement sites, shown on a map in Fig. 1, represent different urban environments and different aspects on the human exposure of particulate matter.

The residential area with detached houses about 16 km west of Helsinki city center (RA suburban, Fig. 1a) and the residential area with densely constructed apartment houses near the Helsinki city center (RA inner city, Fig. 1b) represent different residential areas. In the RA suburban, there were only small roads with light traffic near the measurement station (the closest main road was located at a distance of 1.3 km) and in the RA inner city the measurement station located on a sports field. Thus, at these locations, measurements were not significantly affected by fresh emissions from traffic. However, these locations are important from the point of view of human exposure because people spend time there, and the contribution of outdoor concentrations on the indoor concentrations can also be estimated.

Measurements at the traffic site in the inner city (TS inner city, Fig. 1b) and the traffic site next to a major road (TS major road, Fig. 1a) represent two types of traffic sites. The measurement station in the inner city located in a crossing area of two busy roads, and the other station along one of the main commuting roads of Helsinki area (Ring I). The measurement stations were located approximately at a distance of 2 m from the road at the TS inner city and approximately 5 m at the TS major road. Also the measurement station in the city center (City center, Fig. 1b) could be regarded as a traffic site, because the aerosol sample was taken above a bus stop next to a busy street. The city center and the TS inner city were street canyon type environments whereas the TS major road located at open environment. Yearly averages for the traffic volume were 44,000 vehicles per workday at the TS inner city and 20,000 vehicles per workday in the city center (Mannerheimintie) measured in 2010 (Lilleberg and Hellman, 2011). At the TS major road, the traffic volume was 69,000 vehicles per workday in 2012 according to the road statistics monitored by the Finnish Transport Agency. From the point of view of human exposure, these three locations differ significantly from each other. In the city center, many pedestrians are exposed to particles directly, while at the TS major road and at the TS inner city, the exposure occurs mostly inside vehicles.

During the stationary measurement campaigns, except the city center campaign, the instrumentation was installed inside a movable air quality measurement station (see e.g. Järvinen et al., 2015; Saari et al., 2015). The aerosol sample was taken from the roof of a station without any pre-conditioning of the sample. During the campaign in the city center, the instrumentation was installed into a room below ground level and the aerosol sample was taken directly above. In all the measurements, the tubing from sampling points to measurement instruments was made by stainless steel tubes from which the air was taken to the instruments by Tygon R-3603 tubing (Saint-Gobain). In addition to the main instrument ELPI, also NSAM, DMPS, PM2.5, NO (nitrogen monoxide) and NO_2 (nitrogen dioxide) measurements were conducted. The PM2.5 concentrations were measured using different monitors based on β-attenuation (FH 62 I-R in the city center), combination of light-scattering and β-attenuation (SHARP 5030 in the RA suburban) or with a tapered element oscillating microbalance technique (TEOM 1400 AB in the RA inner city, at the TS inner city and at the TS major road). The PM2.5 monitoring data of different instruments was corrected using calibration equations to ensure equivalent results according to European standards (Waldén et al., 2010). NO and NO_2 concentrations were measured with a chemiluminescence analyzer (Horiba APNA360/370).

2.3. Mobile laboratory measurements

The LDSA size distributions were measured also using the ELPI installed into the mobile laboratory "Sniffer" (Pirjola et al., 2004, 2006, 2012; Lähde et al., 2014). In these measurements, the aerosol sample was taken at a height of 2.4 m from the ground level above the vans windshield. In addition to the LDSA size distributions, the total LDSA concentration was measured using the NSAM. A weather station on the roof of the van at a height of 2.9 m above the ground level provided the meteorological data. The relative wind speed and direction were measured with an ultrasonic wind sensor (Model WAS425AH, Vaisala), the temperature and relative humidity with temperature and humidity probes (Model HMP45A, Vaisala). In addition, a global positioning system (model GPS V, Garmin) saved the vans speed and the driving route.

The mobile laboratory was used to complete the measurement network with suburban residential areas (RA 1–6, Fig. 1a) and a park area near the Helsinki city center (Park area, Fig. 1b). The measurement location in the park area called Hietaniemi was located on the seashore with very clean air. The names of the residential areas were Kattilalaakso (RA 1), Laaksolahti (RA 2), Lintuvaara (RA 3), Vartiokylä (RA 4), Itä-Hakkila (RA 5) and Päiväkumpu (RA 6). The mobile laboratory measurements took place between 15th and 27th February 2012. A certain route was driven in each residential area twice in a day and several times during the measurement period. The measurements consisted of one morning (all)

Table 1
Locations and time periods for the stationary measurement campaigns. Also the average temperature and relative humidity during the campaigns with the standard deviation as an error limit are shown. TS refers to a traffic site and RA to a residential area.

Location	Time period	Temperature (°C)	RH (%)	Area description
Kattilalaakso	14.2.–28.2.2012	−3.0 ± 2.5	82 ± 8.7	RA suburban
Kallio	20.2.–28.2.2012	−2.0 ± 2.5	83 ± 9.5	RA inner city
Mannerheimintie	30.11.–15.12.2010	−6.6 ± 3.7	80 ± 8.8	City center
Töölöntulli	1.12–14.12.2010	−5.8 ± 3.1	80 ± 9.2	TS inner city
Ring I, Malmi	19.10.–7.11.2012	3.8 ± 3.5	78 ± 12	TS major road

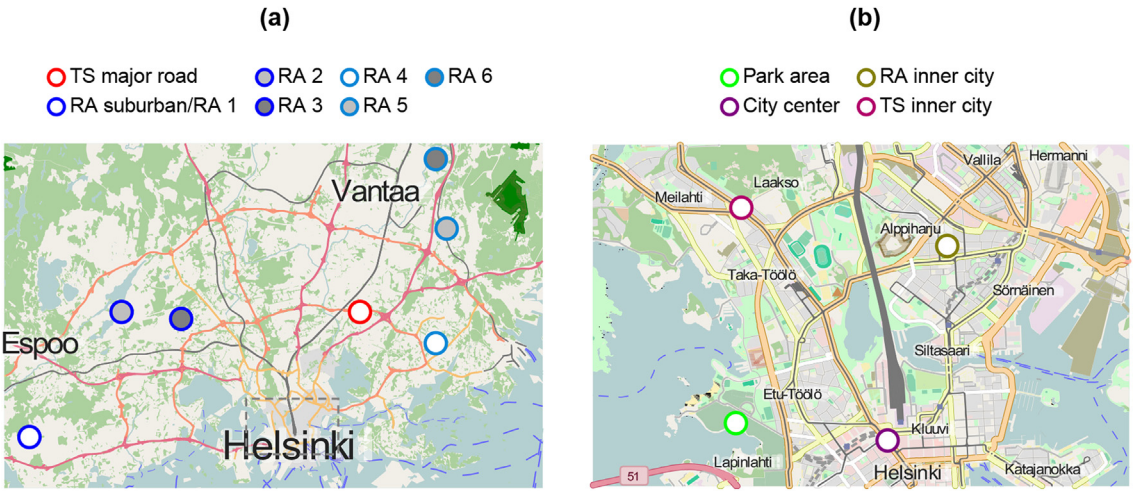

Fig. 1. Measurement locations (a) outside the inner city and (b) in the inner city on the map. TS refers to a traffic site and RA to a residential area. (©OpenStreetMap contributors).

and four (RA 1–3) or five (RA 4–6) evenings for all the residential areas, including approximately 30min data for each. The morning measurements took place between 8 a.m. and 1 p.m. and the evening measurements between 5 p.m. and 10 p.m. In the park area, the mobile laboratory visited in the end of each measurement day. In addition to the data collected in the residential areas and the park area, also some on-road data was collected during transitions between the locations. The on-road data was divided into two groups including major roads and the city center.

3. Calibration

3.1. Lung deposited surface area

The definition of the lung deposited surface area (LDSA) concentration is based on the experimentally defined deposition efficiency of human lungs. The deposition efficiency can be measured separately for tracheobronchial or alveolar region of the human respiratory system. Usually, as in this study, the LDSA is defined for the alveolar region. We used the alveolar deposition efficiency as a function of the particle size presented in a report by ICRP (1994). The NSAM has been calibrated using the same standard for the deposition efficiency (Fissan et al., 2007). In this study, the calibration means the comparison of the instrument responses and the determination of the calibration factor.

Any aerosol device based on diffusion charging can be calibrated to measure the LDSA concentration by comparing the output current to a reference. The reference LDSA concentration can be measured with a reference instrument or calculated from a number mobility size distribution measured with a reference instrument. In the number size distribution based calibration, the reference LDSA concentration can be calculated by multiplying the number size distribution with the deposition efficiency and summing up the total LDSA concentration of the aerosol. Both of these calibration methods were used in this work.

3.2. Mobile laboratory data

The ELPI was calibrated in the mobile laboratory to measure the LDSA concentration by comparing its output to the output of an NSAM. Fig. 2 shows the LDSA measured with the NSAM as a function of the ELPI total current averaged over 1 min. A linear correlation between these two metrics was found with the correlation coefficient value of 0.84, and a calibration factor of 60 $\mu m^2/(cm^3\ pA)$ was obtained from the slope of a linear fit. In this study, we used this value to convert the ELPI current to the LDSA concentration. Thus, the final LDSA size distributions were obtained from the current size distributions by multiplying these with the calibration factor. The strenghts of the mobile laboratory calibration were the large amount of data, uniform sampling lines and the variety of different environments. The calibration factor obtained from the mobile laboratory calibration was very close to the calibration factor value determined similarly by Mäkelä et al. (2009) for titanium dioxide (70 $\mu m^2/(cm^3\ pA)$) and iron oxide (64 $\mu m^2/(cm^3\ pA)$) nanoparticles.

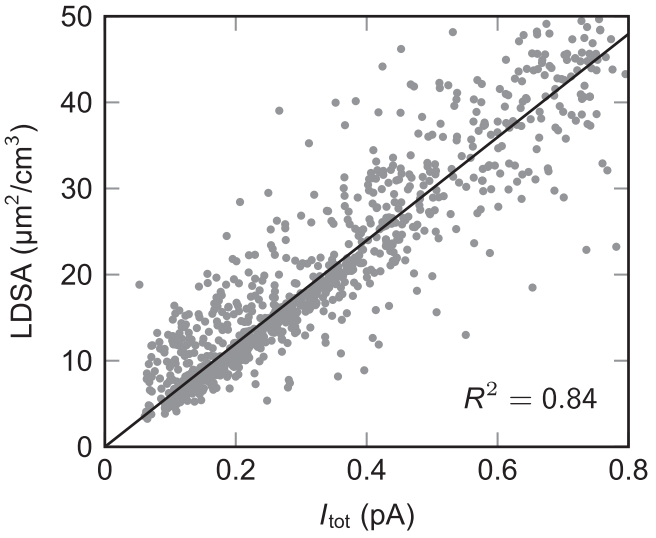

Fig. 2. The lung deposited surface area (LDSA) concentration measured by the nanoparticle surface area monitor (NSAM) as a function of the electrical low pressure impactor (ELPI) total current for mobile laboratory measurements in different environments. The data is averaged over 1 min. The slope of the fit and the calibration factor was determined to be 60 $\mu m^2/(cm^3\ pA)$. Also the correlation coefficient (R^2) value for the data is seen.

3.3. Stationary measurement data

To ensure the capability of the ELPI to measure the LDSA concentration, additional calibrations were conducted using the stationary measurement data. Fig. 3 shows the LDSA concentration

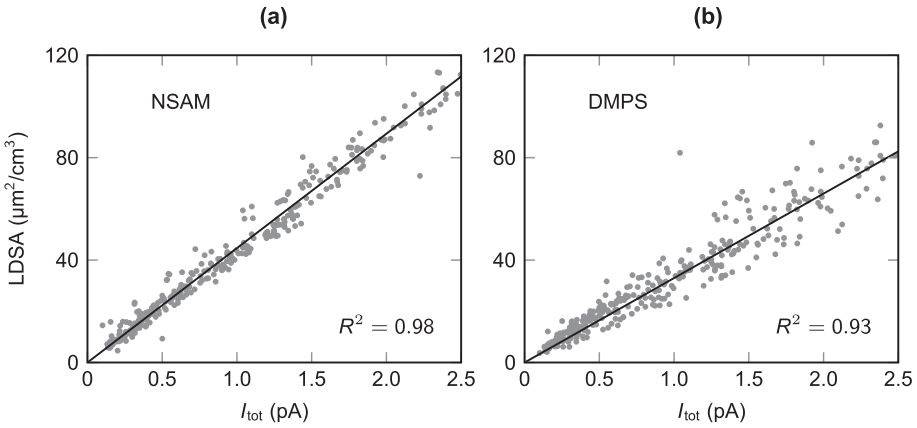

Fig. 3. The lung deposited surface area (LDSA) concentration (a) measured by the nanoparticle surface area monitor (NSAM) and (b) calculated from the differential mobility particle sizer (DMPS) data as a function of the electrical low pressure impactor (ELPI) current for stationary measurements at the traffic site next to the major road. The data is averaged over 1 h. The slopes of the fits were 43 and 33 $\mu m^2/(cm^3$ pA) for the NSAM and DMPS, respectively. Also the correlation coefficient (R^2) values for the data are seen.

measured by the NSAM and calculated from the DMPS number size distributions as a function of the ELPI total current. This data was measured at the traffic site next to the major road and averaged over 1 h. Both of these cases showed a linear correlation with relatively high correlation coefficient values (0.98 and 0.93), which approves the capability of the ELPI to measure the LDSA concentration. However, the calibration factors, 43 and 33 $\mu m^2/(cm^3$ pA) for the NSAM and DMPS, respectively, differed from the calibration factor obtained for the mobile laboratory data. This difference can partly be explained by the fact that different environments emphasize different particle size ranges in the LDSA. At the traffic sites, the small nucleation mode particles are more important than in the residential areas. As shown above, the mobile laboratory calibration covered all the different environments and the response was still linear. The correlation in the stationary data at the traffic site was even better, but the calibration factors could include errors affecting the absolute values and they were not comprehensive with respect to different environments. Some of the errors affecting the absolute values could be the differences in the inlets and sampling lines for different instruments, combined with the extremely fresh traffic related aerosol and its fast changes. In general, small errors in the flows of the DMPS may result in a significant error in the total concentration, which could also cause error in the calibration factor value. The NSAM and the ELPI with one major sample flow are more reliable in this respect.

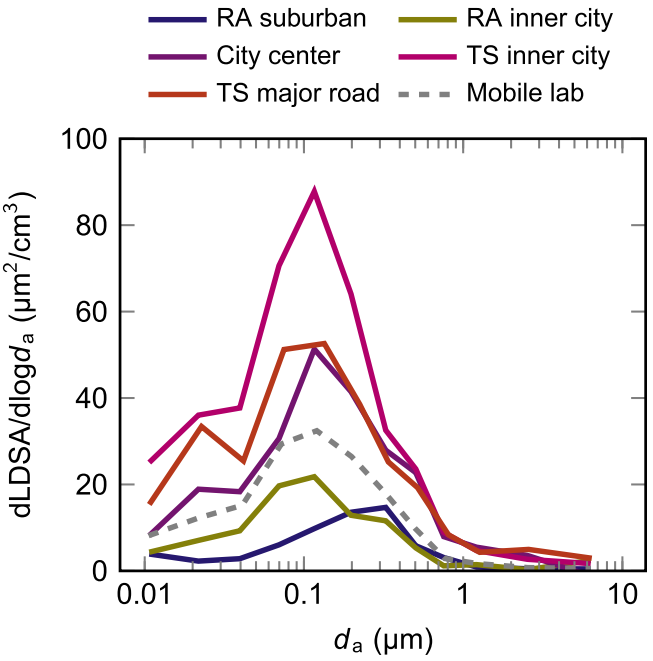

Fig. 4. The averaged lung deposited surface area (LDSA) size distributions from the stationary measurements. Also the mean LDSA size distribution from the mobile laboratory measurements is shown. TS refers to a traffic site and RA to a residential area.

4. Results and discussion

Fig. 4 shows the LDSA size distributions from the stationary measurements averaged over the measurement periods. At the traffic sites, two-modal size distributions were clearly distinguished. The soot mode dominated but also the nucleation mode had a clear contribution to the size distribution and the total LDSA concentration. These two modes were also distinguished in the city center where the instant influence of traffic was seen. However, compared to the traffic sites, the soot mode was peaking at a larger particle size and the concentration of the nucleation mode was lower. This indicates that the aerosol was on average more aged but still its main source was traffic. In the LDSA size distribution of the RA inner city, the total concentrations were clearly lower than at the traffic sites and in the city center. The role of the nucleation mode was not significant but the influence of traffic and the two modes were still seen. In the RA suburban, the LDSA concentrations were lower and the LDSA size distribution had shifted to larger particle sizes indicating an influence of aged traffic-related aerosol or wood burning. Also the influence of long-range transported aerosol was more important than at the traffic sites.

In Fig. 5, the LDSA size distributions from the mobile laboratory measurements have been averaged and sorted according to the measurement location. The size distributions representing mobile laboratory measurements on major roads and in the city center were very similar to the size distributions obtained from the stationary measurements at the traffic sites and in the city center, respectively. This indicates that the stationary measurements represented well such a type of an urban environment. Fig. 5 also shows the LDSA size distributions measured in the park area and in the six different suburban residential areas with detached houses. The concentrations were much lower than in the environments clearly influenced by traffic. In the park area, the LDSA size

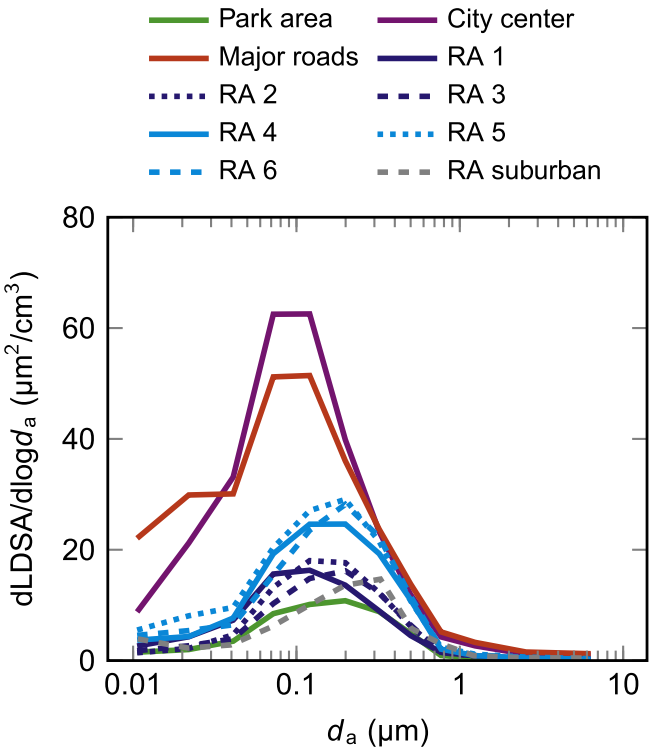

Fig. 5. The averaged lung deposited surface area (LDSA) size distributions from the mobile laboratory measurements in different environments. RA refers to a residential area. Also the average size distribution for the stationary measurements in the RA suburban (corresponding to RA 1) is shown.

distribution was dominated by the long-range transported aerosol that most likely originated from Eastern Europe. This could be seen in the variation of different measurement days and confirmed by comparing the simultaneous particulate measurements around the city and elsewhere. Also in the residential areas, the influence of the long-range transported aerosol was significant. During the measurements in the first three residential areas (RA 1–3), the background formed by the long-range transported aerosol was much lower than during the measurements at the rest of the areas (RA 4–6). In addition to this background, local sources, such as wood burning and to some extent also traffic, could have had an effect on the LDSA size distributions.

Table 2 shows the average total concentrations of the LDSA in different environments for both the stationary and mobile laboratory measurements. The highest concentration of 94 μm^2/cm^3 was measured at the TS inner city. The other environments close to traffic sources had concentrations between 53 and 67 μm^2/cm^3. In the park area, the average LDSA concentration was only 12 μm^2/cm^3, which can be considered relatively low compared to the previously measured values (10–89 μm^2/cm^3) at urban background sites (Reche et al., 2015; Wilson et al., 2007; Fierz et al., 2011; Ntziachristos et al., 2007; Gomes et al., 2012). The RA inner city had approximately the same average LDSA concentration of 23 μm^2/cm^3 as all the residential areas outside the inner city on average, although there were clear differences in the size distributions, as shown previously. The average concentration obtained from the stationary measurements in Kattilalaakso (RA suburban) was lower than the concentration obtained from the mobile laboratory measurements (RA 1), mainly because the stationary measurements were running also during night time. Altogether, the average LDSA concentrations at the traffic sites and in the city center were lower than the previously reported concentrations at traffic sites in Los Angeles (106–153 μm^2/cm^3) reported by Ntziachristos et al. (2007).

An idea of the temporal deviation of the LDSA concentrations is given by the 10th and 90th percentiles in Table 2. The 90th percentiles show that, relatively often, the concentrations could be at the traffic sites and in the city center above 100 μm^2/cm^3, even as high as 200 μm^2/cm^3. On the other hand, the 10th percentiles were on the level of urban background, except the mobile measurements in the city center. The smallest temporal deviations of the LDSA were seen in the RA suburban and in the park area. Also the average concentrations of the PM2.5, NO and NO_2 for the stationary measurements are seen in Table 2. The concentrations of NO and NO_2 were higher at the traffic-related environments as expected. An interesting finding was that the average PM2.5 in the RA suburban was at the level of the most congested traffic site (TS inner city), even though the LDSA concentration was over 6 times lower. This indicates that the local sources around the RA suburban, such as wood burning, seem to contribute significantly to the PM2.5 but not for the LDSA.

To observe the relation of the LDSA concentration to the commonly measured air pollution parameters, Fig. 6 shows the LDSA concentration as a function of the PM2.5, NO and NO_2 for the stationary measurements. The data was averaged over 1 h. Linear

Table 2
The average total lung deposited surface area (LDSA) concentrations in different environments for both the stationary and mobile laboratory measurements. The deviation of the concentrations is described by the 10th and 90th percentiles. Also the average concentrations of the PM2.5, NO and NO_2 for stationary measurements are shown. TS refers to a traffic site and RA to a residential area.

Area description	Location	LDSA	LDSA percentiles		PM2.5	NO	NO_2
		(μm^2/cm^3)	10th	90th	(μm^2/cm^3)	(μm^2/cm^3)	(μm^2/cm^3)
TS inner city	Töölöntulli	94	13	203	11.4	12.7	33.0
TS major road	Malmi	67	15	142	9.6	50.3	43.8
City center	Mannerheimintie	54	15	101	10.1	13.6	33.3
RA inner city	Kallio	23	8	40	4.3	4.8	22.6
RA suburban	Kattilalaakso	15	7	24	11.3	5.2	24.3
Park area	Hietaniemi	12	4	23	–	–	–
Major roads	–	65	14	123	–	–	–
City center	–	64	24	119	–	–	–
RA 1	Kattilalaakso	17	5	28	–	–	–
RA 2	Laaksolahti	17	8	26	–	–	–
RA 3	Lintuvaara	16	7	26	–	–	–
RA 4	Vartiokylä	26	7	59	–	–	–
RA 5	Itä-Hakkila	31	7	61	–	–	–
RA 6	Päiväkumpu	27	7	61	–	–	–

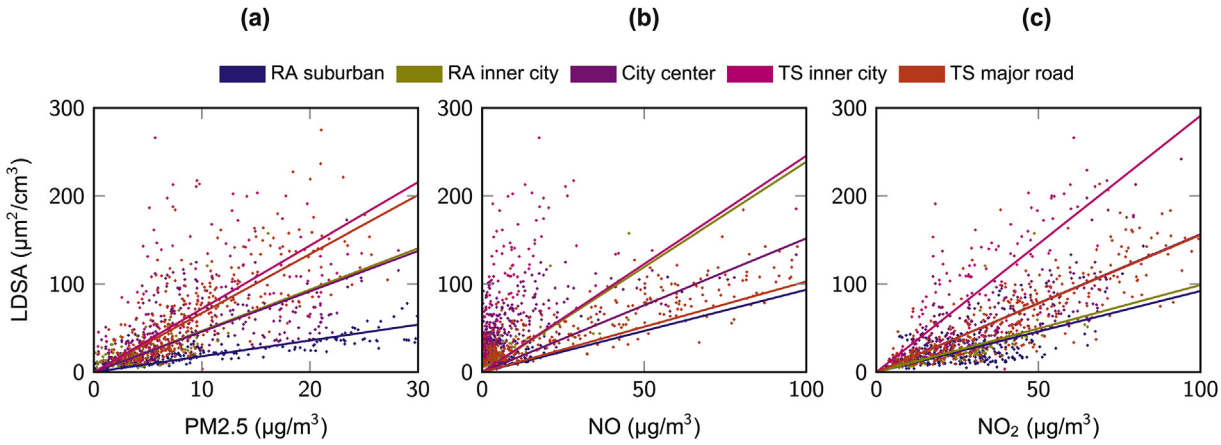

Fig. 6. The LDSA concentration as a function of (a) the PM2.5, (b) NO and (c) NO_2 for the stationary measurements. The data was averaged over 1 h. The linear fits show the correlation between these metrics, separately for the different environments. TS refers to a traffic site and RA to a residential area. The slopes of the fits with the correlation coefficients are seen in Table 3.

functions were fitted into the data separately for the different environments, and the obtained slopes with the correlation coefficients (R^2) can be seen in Table 3. The LDSA correlated with the nitrogen dioxide well and the slope clearly increased with the influence of traffic (Fig. 6c). The smallest correlation was observed for the RA suburban, which indicates the influence of other emission sources than traffic on the LDSA. The correlation between the LDSA and nitrogen monoxide was negligible (Fig. 6b), but the two measures of particulate matter, the LDSA and the PM2.5, correlated with each other well in most of the environments (Fig. 6a). However, there was a significant variation in the slopes of the fits from 1.8 to 7.2 $m^2/\mu g$, measured in the RA suburban and at the TS inner city, respectively. The slope values increased very consistently with the increasing influence of traffic. This result indicates that, even if the PM2.5 would be high in the suburban residential areas, the LDSA may be relatively low compared to the traffic-related environments. The slope values obtained in this correlation analysis can be used to roughly estimate the LDSA concentration in different environments based on the PM2.5 measurements.

Fig. 7 shows the averaged and normalized size distributions for the mass, number and LDSA measured at the traffic site next to the major road. The mass size distribution was calculated from the DMPS data assuming unit density. The three uppermost channels were removed from the DMPS surface area and mass size distributions due to a measurement artefact. In the number size distribution, the nucleation mode clearly dominated, and the mass size distribution mainly consisted of the soot mode. However, in the LDSA size distribution, both the nucleation mode and the soot mode were distinguished and they both contributed to the total LDSA concentration. In all of these size distributions, also other modes, such as the accumulation mode of the background aerosol, could be there, but the fresh traffic-related modes clearly dominated. Fig. 7 also shows the usual range of the most penetrating particle size (MPPS) for aerosol filtration (Podgórski et al.,

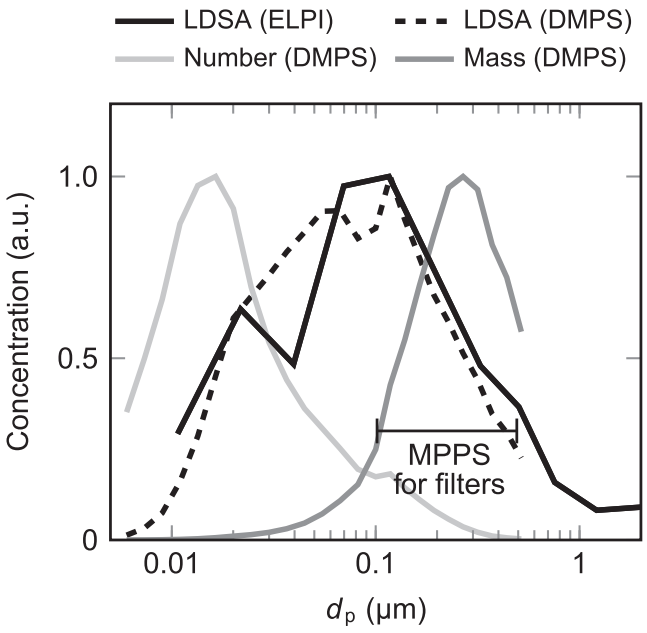

Fig. 7. The normalized number and mass size distributions compared to the normalized lung deposited surface area (LDSA) size distribution from the stationary measurements at the traffic site next to the major road. The distributions were averaged over the measurement period. Also the range of the most penetrating particle size (MPPS) for filters is shown.

Table 3
The slopes and correlation coefficients from the comparison of the LDSA to the PM2.5, NO and NO_2 for the stationary measurements. TS refers to a traffic site and RA to a residential area.

Area description	Location	PM2.5		NO		NO_2	
		Slope ($m^2/\mu g$)	R^2	Slope ($m^2/\mu g$)	R^2	Slope ($m^2/\mu g$)	R^2
TS inner city	Töölöntulli	7.2	−0.01	2.5	−1.59	2.9	0.49
TS major road	Malmi	6.7	0.58	1.0	0.64	1.6	0.73
City center	Mannerheimintie	4.6	0.23	1.5	−0.46	1.6	0.57
RA inner city	Kallio	4.7	0.24	2.4	−0.12	1.0	0.44
RA suburban	Kattilalaakso	1.8	0.58	0.9	−0.73	0.9	0.32

2006). It is seen that a significant part of the mass and LDSA size distributions are at this size range but only a small fraction of the number size distribution. In general, the contribution of outdoor concentrations on the concentrations inside buildings and vehicles is dependent on the particle size through penetration factors. In order to estimate the indoor concentrations, the size distribution and the size dependent penetration must be known.

5. Conclusions

In this study, the lung deposited surface area (LDSA) size distributions were measured in different urban outdoor environments in the metropolitan area of Helsinki. Two-modal size distributions were clearly distinguished at the traffic sites and in the city center. It means that the both traffic related particle modes, the nucleation mode and the soot mode, had a clear contribution to the total concentration of the LDSA in urban environments. The influence of traffic could also be seen in the LDSA size distribution measured at the residential area in the inner city. In the suburban residential areas, the LDSA size distributions were more affected by the long-range transported background aerosol and other sources, such as the local wood burning. The average total concentrations of the LDSA varied from 12 to 94 $\mu m^2/cm^3$. The lowest average concentration was measured in the park area and the highest concentration at the traffic site next to the major road.

The LDSA concentration has been shown to correlate with negative health effects in toxicological studies. However, the availability of the total concentrations of the LDSA in urban areas is limited in the literature. The results of this study provide valuable information on the LDSA concentrations in different environments inside the same metropolitan area. This information can be used for the purpose of epidemiological studies and urban planning. One of the most interesting results of this study was that the mass of fine particles (PM2.5) correlated with the LDSA, but the relation of these two metrics depended on the type of the environment. Consequently, the same amount of particulate matter in terms of PM2.5 can be over three times more harmful for human health near traffic than in suburban residential areas, if the LDSA concentration is considered as a right metric for the negative health effects.

The LDSA size distributions in urban areas have not been reported in any previous studies. The main instrument in this study was the electrical low pressure impactor (ELPI), which was calibrated and used to measure the LDSA. The advantage of the ELPI, compared to many other instruments capable of measuring the LDSA, is that it also measures the size distribution of the LDSA. The calibration factor, obtained in this work by comparing the instrument response to the reference instruments, can also be used for other studies to convert the ELPI current signal to the LDSA. The average size distributions reported in this work provide a lot of information in addition to the total concentrations. The size distributions can reveal, for instance, the ageing and origin of the aerosol. In addition, by using the size distribution data, it is possible to estimate the contribution of the LDSA outdoor concentration on the concentrations inside buildings and vehicles through size-dependent penetration factors. Simple hand-held instruments measuring the total LDSA concentration, such as the NanoTracer and the Partector, will most likely become more common in the near future. Along with the data provided by these instruments, it is also important to measure the size distributions in order to cover different outdoor and indoor environments for the use of epidemiological studies and exposure estimations.

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